Journal of Carbon Research

Article

Laser-Induced Refractive Index Indicates the Concurrent Role of the Bio-Structuration Process in the Comparison with the Nano-Structuration One

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Abstract: It should be remarked that the basic knowledge collected from complicated area of the structuration process of the organic materials, including the liquid crystal (LC) ones, useful for the optoelectronics and biomedicine, requires extending the types of the novel matrix model materials and the class of the dopants, which can change the spectral and photorefractive features of the matrixes with good advantage. In the current paper the effect of the introduction of the bio-objects (based on DNA) and of the nano-objects (based on fullerenes, quantum dots, carbon nanotubes, shungites, graphenes) in the organic conjugated materials has been comparatively discussed. The influence of this process on the photorefractive features, namely on the laser-induced change of the refractive index, has been studied. The clear innovative tendency of the alternative using of the bio-objects together or instead of the nano-objects ones has been analyzed via considering of the modification of the spectral and non-linear optical characteristics.

Keywords: organic materials; laser-induced change of the refractive index; doping process; DNA; fullerenes; quantum dots; carbon nanotubes; shungites; liquid crystal; laser-mater interaction

Citation: Kamanina, N.; Likhomanova, S.; Zubtsova, Y. Laser-Induced Refractive Index Indicates the Concurrent Role of the Bio-Structuration Process in the Comparison with the Nano-Structuration One. *C* **2022**, *8*, 43. https://doi.org/10.3390/c8030043

Academic Editor: Giuseppe Cirillo

Received: 2 August 2022
Accepted: 22 August 2022
Published: 26 August 2022

1. Introduction

Last two decades the organic thin-film materials can be considered as the alternative ones in the comparison with the volumetric inorganic compounds used in the different area of the telecommunications, laser technique, display and biomedicine. It should be mentioned that many methods and processes have been applied in order to modify the optoelectronic properties of the organics. Among them the structuration via the application of the nano- and bio-objects doping occupies the special place. From this point of view, the basic knowledge collected from the features of the structuration process of the organic materials, including the liquid crystal (LC) ones, can be useful for the optoelectronics and biomedicine. Indeed, it requires extending the types of the novel matrix model materials and the class of the dopants, which can change the spectral and photorefractive features of the matrixes. Different scientific groups work now with the nano-objects-doped [1–8] and bio-objects-doped [9–11] organic composites finding the modification of the structural, spectral, dynamic, acousto-optic, photoconductive and photorefractive parameters. It should be taken into account that one of the mechanisms responsible for the innovative property of the materials appearing coincided with the inter-molecular charge transfer complex (CTC) formation. Incorporated nano- and bio-particles provoke the self-arrangement effect, change of the order parameters, and create the network that dramatically improves the physic-chemical characteristics of the organics.

It should be remarked as well that so many technical approach and set-up can be used to predict the novel properties of the materials, including the doped ones. All of them try to find the correlation between the basic materials features in order to answer the question: What the better area of the materials application for the human life and specific device use? For example, the novel materials can be used in the spatial light modulators based on the liquid crystals [12–15] to write-read the information, to convert them, etc.; the new materials can be applied as the temperature meter, as the innovative sensors, as the system for the photothermal cancer therapy, as the membranes, etc. [16–20].

Our own studies have established that the structural, spectral, nonlinear optical and photoconductive parameters have been modified dramatically [4,11,21–32]. It should be remarked that the IR-spectral shift for the sensitized organics have been found too.

It is clear that one should make the comparative analysis of the nano- and bio-objects influence on the dynamics, photoconductivity and photorefractivity due to the reason that, firstly, the reserve of the synthetic nanoparticles can be exhausted and, secondly, nanoparticles often exhibit the toxicity that can negatively effect on their use in the biophysics and biomedicine.

Based on the change of the spectral and photorefractive parameters of the sensitized organic materials in the current paper it is considering the opportunity of the alternative using of the bio-structuring process together or instead of the nano-structuring one. From this point of view is the modification namely of the photorefractive parameters no doubt. Exactly, the change of the photorefractivity via nonlinear optical study correlated with the dynamics (due to change of the dipole polarizability and increase of the absorption cross section) and with the photoconductivity characteristics (due to the increase of the barrier-free pathway for the electrons and via change of the charge carrier mobility). In this case the estimation of the refractive parameters such as the laser-induced change of the refractive index and the nonlinear refraction as well as the cubic nonlinearity can be made with good advantage. At that, one should testify, that to analyze the nonlinear optical processes it should be taken into account that when the electric field of the laser wave is less than the intra-atomic electric field correlated with the electron charge and with the Bohr radius, we should estimate the linear effect. But, when the electric field of the electromagnetic wave is larger than the intra-atomic electric field, we should draw the attention on the nonlinear optical features. Using this aspect, the values of the optical susceptibilities play an important role in the nonlinear optical effect. Really, the most important optical characteristic of the all inorganic or organic materials with different symmetry is the induced dipole, which can be expressed through the dipole polarizabilities $\alpha^{(n)}$. These last values are correlated with the nonlinear susceptibility $\chi^{(n)}$ and the local volume υ of the materials. Thus, the laser-matter interaction provokes the change in the polarization of the media and predicts the change of the important properties, such as dynamic, photorefractive and photoconductive ones. Moreover, it can testify the correlation between the parameters mentioned above.

In the current paper it can be shown the analogous refractive parameters of the bio-objects doped organics in comparison with the nano-objects-doped ones.

2. Materials and Methods

As has been accented in the previous paragraph the photorefractive characteristics responsible for the relationship with the other basic parameters and correlated with other materials features have been chosen as the basic characteristics to study. The photorefractive properties have been investigated using the model organic systems with the intra-molecular CTC due to initial donor-acceptor interaction. They can be as following conjugated organics: polyimide (PI), pyridine (for example, 2-cyclooctylamine-5-nitropyridine—COANP) compounds as well as the nematic liquid crystals (NLC) from cyanobiphenyl group. As the effective intermolecular dopants and efficient electron acceptors the fullerenes, shungites, reduced graphene oxides, and quantum dots (QDs), as well as the bio-components, namely, red fish DNA have been used. The thin films of the organics sensitized with the nano-objects have been prepared with the thickness of 2–5 micrometers. Nano-objects concentration *c* was varied in the range of 0.003–5 wt.%. The fullerenes and carbon nanotubes (CNTs) have been purchased from Alfa Aesar (Kurlsruhe, Germany); shungite structures have been produced by Karelian Research Centre RAS. The thickness of the LC cells with conducting ITO electrodes deposited on the glass substrates was 10 micrometers. Water solution of the DNA with the concentration of 4.72 g/L has been used. The relation between the LC mixture and the DNA water solution was ~5:1. It should be noticed that the linear refractive index of the compounds studied was measured by the ellipsometer Horiba Jobin Yvon Uvisel.

It should be taken into account, that the conducting layers of the LC-cells have been modified using the surface electromagnetic wave (SEW) via the method shown before [33,34]. In this case, the conducting ITO coatings perform two functions, namely, it operates as the orienting relief for the LC-dipole alignment and as the conductor. Moreover, to increase the transparency of the ITO coatings the carbon nanotubes with the refractive index that is close to 1.1 have been deposited and treated by the SEW too.

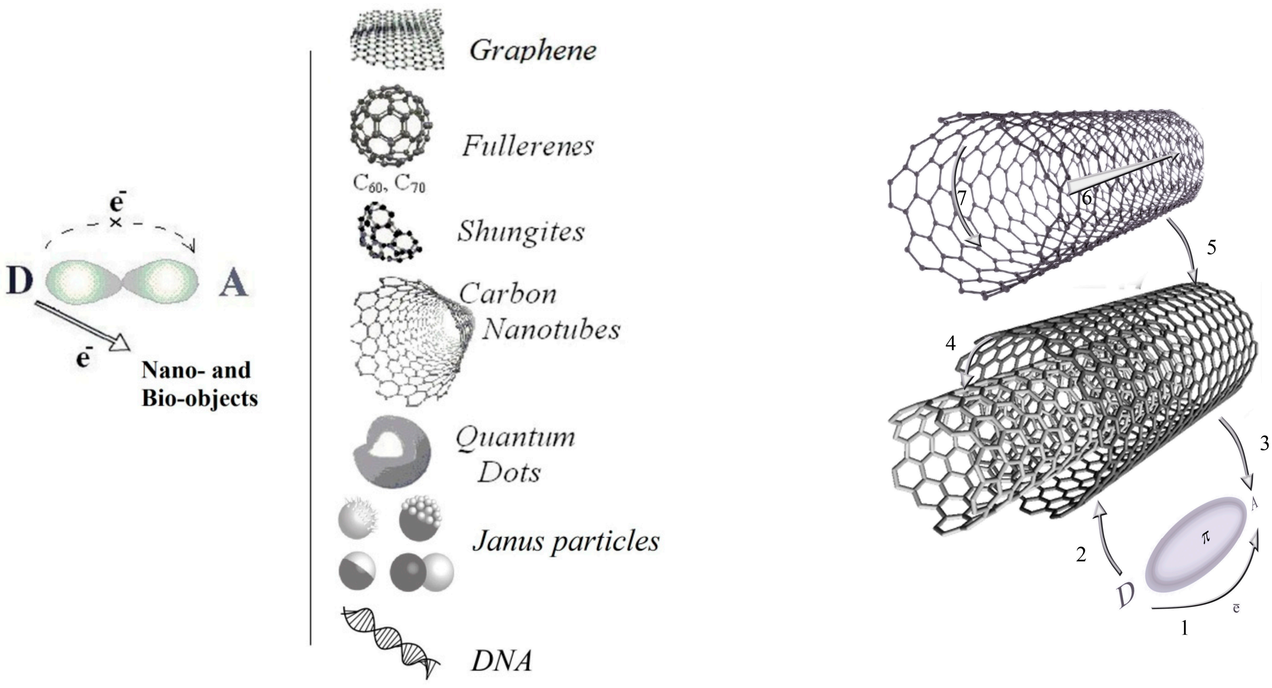
The second harmonic of the pulsed nanosecond Nd-laser at wave length of 532 nm has been used. The laser energy density W has been chosen in the range of 0.03×10^{-3}–$0.6\ \mathrm{J} \times \mathrm{cm}^{-2}$. The pulse width of 10–30 ns has been applied. The amplitude-phase thin gratings have been recorded under the Raman-Nath diffraction conditions analogous to that shown previously in papers [35,36] at spatial frequency Λ of 90–150 mm^{-1}. We have applied the holographic recording technique to activate the reversible grating at $\Lambda^{-1} \geq d$, where Λ is the spatial frequency, d is the thickness of the film. The laser-induced change of the refractive index for the media with the large value of the cubic nonlinearity has been estimated via dependence of the diffraction efficiency on energy density. The mathematical procedure to calculate the nonlinear refraction coefficient n_2 and cubic nonlinearity $\chi^{(3)}$ has been proposed in the ref. [37], then has been firstly used for the fullerene-doped liquid crystal organics in ref. [26] and has been there after extended for the different types of the nanostructured materials in the papers [11,26,28,29].

It should be taken into account that laser technique, especially holographic scheme has been classically often used by the material science area researchers. Organics based on thin-film polyimide materials; liquid crystal compounds, etc. have been treated via this technique, Z-scanning scheme or via the third harmonic generation approach. Photorefractivity change has been found under the different conditions of the input energy densities [38–49]. We have used the holographic set-up due to the fact that this method permits to find the change of the refractivity in the reversible mode.

3. Results and Discussion

Let us to repeat the role of the refractivity in the advances in the material nanosensitization. Really, as has been shown and postulated in paper [50], refractive property changes can be considered as the main parameter to indicate the organic material physical–chemical feature improvements. In order to adequately understand of the idea of the bio-objects alternative using instead or together with the nano-objects ones via estimation of the organics photorefractivity changing, let see Figure 1.

It should be remarked that all systems considered are the materials with the initial intra-molecular CTC formation process. Moreover, the initial acceptor fragment, for example of the polyimide molecule is close to 1.14–1.4 eV; the same value for the COANP molecule is close to 0.45 eV [51]. Under the sensitization process the charge transfer reveals from the intra-molecular donor fragment of the organic conjugated molecules not to its acceptor fragment but to the nano-objects if the electron affinity energy of the nano-objects is larger than the one for the intra-molecular acceptor fragment. It should be mentioned that the electron affinity energy of the fullerene C_{60} and C_{70} is 2.65 and 2.7 eV, respectively. Moreover, as has been shown in the paper [3], fullerene, for example, can accept not one but 6 electrons. Thus, via inter-molecular CTC the creation of the larger dipole moment μ_{inter} can be really possible. This dipole moment μ_{inter} based on the inter-molecular CTC is essentially larger than the one (μ_{intra}) obtained from the intra-molecular process and can be increased up to one order of magnitude. The first supporting experimental result devoted to the measurement of the dipole moment in the pure and the sensitized composite based on the polyimide has been obtained previously and shown in paper [52] and supported in paper [23]. The qualitative model added with the DNA bio-objects shown in Figure 1a is in good coinciding with this fact. Moreover, schematic diagram of the possible charge transfer pathways depending on the arrangement of the introduced intermolecular acceptor relative to the intramolecular donor is presented in Figure 1b for the complicated CTC with the CNTs. The different ways for the electrons are numbered from 1 to 6. It is worth considering the interaction between the *intra*molecular donor of the matrix system and the *intra*molecular acceptor (1), between the *intera*molecular donor and CNT (2), between the *inter*molecular acceptor and CNT (3), since the latter can exhibit both the donor and the acceptor properties. It is also necessary to take into account the charge transition inside the CNT (4), between them (5) and along the CNT (6).



(**a**) (**b**)

Figure 1. (**a**)—Possible qualitative model, which can visualize the charge transfer pathway change for the conjugated nano- and bio-objects-doped organics under the condition when inter-molecular CTC can dominate the intra-molecular one; (**b**)—Schematic diagram of possible charge transfer pathways depending on the arrangement of the introduced intermolecular acceptor relative to the intramolecular donor.

Possibly, the analogous scheme can be used for the DNA-structures of the donor-acceptor systems.

Thus, it can predict the increase of the linear refractive parameters as well (see Figure 2). Measurement of the linear refraction coefficient n by the ellipsometer is presented. The change of the refractive index shows the dramatic increase of it for the LC doped with the bio-objects based on the DNA in the comparison with that obtained for the LC doped with nano-objects.

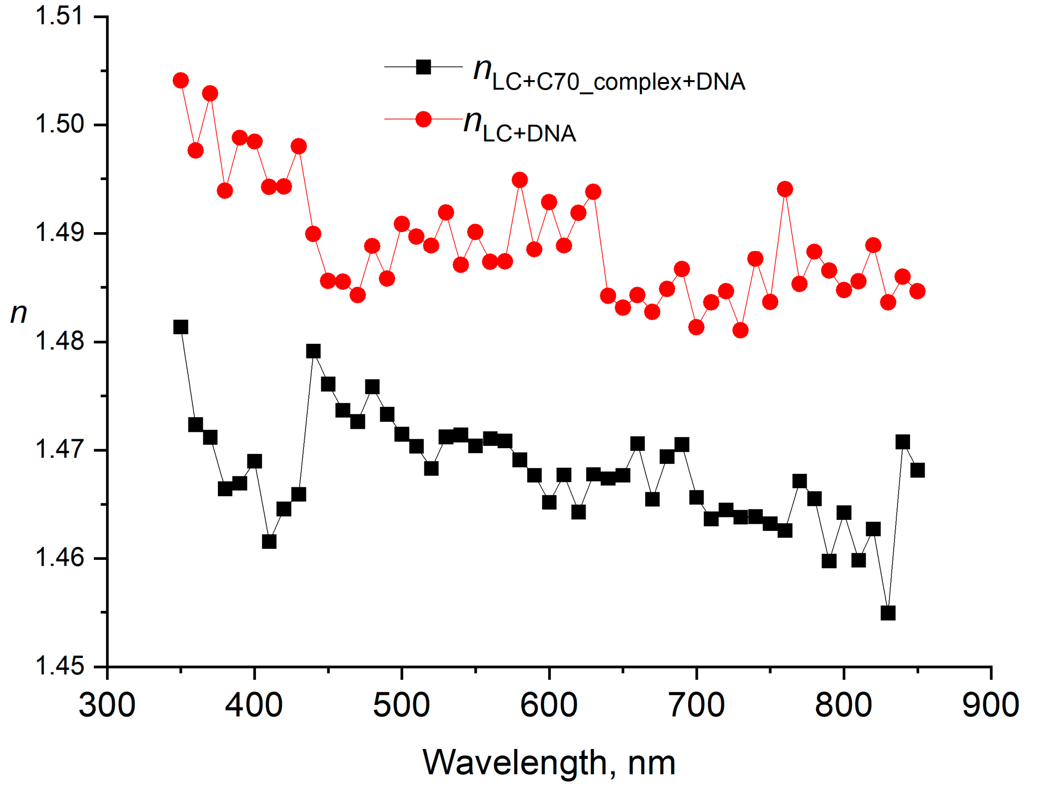


Figure 2. The linear refractive index versus the wavelength for the nano-objects doped materials (dark curve), for the LC doped with the bio-objects doped based on the DNA (red curve).

Analyzing the data shown in Figure 2 it should be mentioned, that classical ellipsometry method can correlate with the laser-induced change of the refractive index. Really, the spectral dependence of the refractive index measured for the liquid crystal mesophase doped with different CTC based on the nano- and the bio-particles testify the increase of the refractive index in the bio-objects doped LC mixture in comparison with the nano-doped one.

Let us to continue to discuss the refractive properties. Indeed, the study of the linear refraction can be correlated with the nonlinear refractive values testing. Data shown in Table 1 can visualize the established data in the laser induces refractivity.

Table 1. Laser-induced change of the refractive index.

System Studied	*c*, wt.%	*W*, J × cm^{-2}	Λ, mm^{-1}	Δn_i	Ref.
Pure PI	0	0.6	90	10^{-4}–10^{-5}	[29]
PI + QDs CdSe(ZnS)	0.003	0.2–0.3	90–100	2.0×10^{-3}	[50]
PI + graphene oxide	0.1	0.2	100	3.4×10^{-3}	[53]
PI + graphene oxide	0.2	0.2	100	3.7×10^{-3}	current
PI + shungite	0.1	0.6	100	3.6×10^{-3}	[50]
PI + shungite	0.2	0.6	100	5.1×10^{-3}	current
PI + C_{60}	0.2	0.5–0.6	90	4.2×10^{-3}	[29]
PI + C_{70}	0.2	0.6	90	4.68×10^{-3}	[54]

Table 1. *Cont.*

System Studied	c, wt.%	W, J × cm^{-2}	Λ, mm^{-1}	Δn_i	Ref.
PI + CNTs	0.1	0.6	100	5.67×10^{-3}	current
PI + CNTs	0.05	0.3	150	4.5×10^{-3}	[31]
PI + CNTs	0.1	0.3	150	5.5×10^{-3}	[31]
Pure COANP	0	0.9	90–100	~10^{-5}	[29]
COANP + C_{60}	5	0.9	100	6.21×10^{-3}	[29]
COANP + C_{70}	5	0.9	100	6.89×10^{-3}	[29]
COANP + graphene oxides		0.2–0.6	100	0.95×10^{-2}	Current
LC based on the complex PI-C_{70}	0.1	0.3	100	1.15×10^{-3}	[50]
LC based on the complex PI-C_{70}	0.2	0.2	90–100	1.2×10^{-3}	Current
LC based on the complex COANP-C_{70}	5	0.02	100	1.4×10^{-3}	[29]
LC with DNA	*	0.1	120	1.39×10^{-3}	[55]
LC based on the complex CdSe(ZnS)-DNA	**	0.1	120	1.35×10^{-3}	[55]
LC based on the complex CdSe(ZnS)-DNA		0.1	100	1.42×10^{-3}	Current
LC	***	0.2 W × cm^{-2}		0.16×10^{-3}	[56]

* Relation between LC and DNA water solution was 5:1 (content of the DNA in water was ~4.72 g × l^{-1}). ** Content of the QDs in nematic was ~0.003 wt.%. *** Data of the paper [34] at the wavelength of 514.5 nm.

Furthermore, the data presented in Table 1 support the influence of the inter-molecular CTC on the change of the laser-induced refractive index Δn_i that correlated with the local volume polarizability changing. Really, to estimate Δn_i the diffraction efficiency η in the first diffraction order has been measured. The relation shown in the Equation (1) has been used to estimate the refractive coefficients based on the mathematical procedure presented in [37].

$$\eta = \frac{I_1}{I_0} = \left(\frac{\pi \Delta n_i d}{2\lambda}\right)^2 \tag{1}$$

where Δn_i is the induced change in the refractive index, I_1 is the intensity in the first diffraction order, I_0 is the input laser intensity, d is the thickness of the medium, λ is the wavelength of the light incident on the medium. After that the nonlinear refraction coefficient n_2 and cubic nonlinearity $\chi^{(3)}$ have been calculated via Equations (2) and (3):

$$n_2 = \frac{\Delta n_i}{I} \tag{2}$$

$$\chi^{(3)} = \frac{n_2 n_0 c}{16\pi^2} \tag{3}$$

It should be analyzed and mentioned that the nonlinear optical coefficients for the nano- and the bio-objects doped organics are close to each other and can be found in the following range:

$$n_2 = 10^{-10} - 10^{-9}\ \text{cm}^2 \times \text{W}^{-1} \text{ and } \chi^{(3)} = 10^{-10} - 10^{-9}\ \text{cm}^3 \times \text{erg}^{-1}$$

These refractivity values can be compared with that estimated for the Si-based materials and for the volumetric $LiNbO_3$ crystals. Moreover, the value of the cubic nonlinearity is close to the one ($\chi^{(3)} = 8.5 \times 10^{-8}$) estimated for the pure nanotubes, shown in patent [57]. Thus, plastic polymer or liquid crystal materials doped with the nanotubes with the little content can replace the pure nanotubes for the different applications. It can predict the protection of the researchers from some toxicity of the carbon nanotubes. Furthermore, the estimated value of the cubic nonlinearity can be compared with that obtained for the classical phthalocyanine materials [58–60]. The place of the materials studied is shown in Figure 3. But, Si-based and lithium niobate structures are the volumetric one, the organic compounds are the thin-film materials.

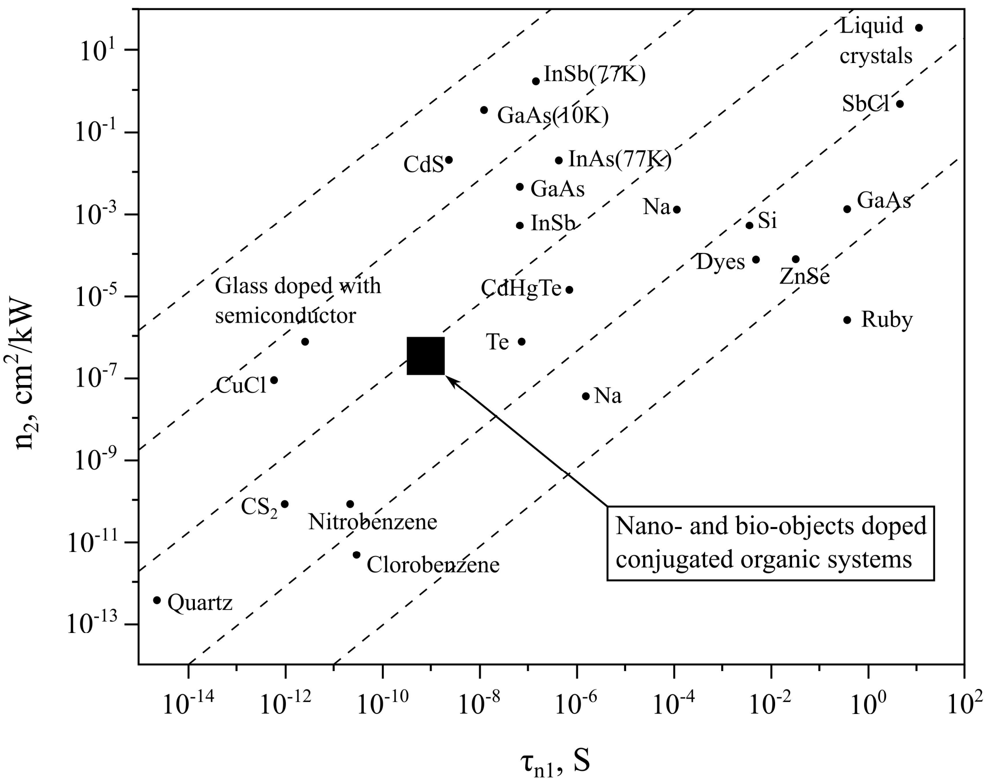


Figure 3. The place of the nano- and bio-objects doped materials among other classical systems with essential refractive features.

Moreover, it shown that bio-objects-doped materials reveal the nonlinear optical coefficients close to that for the nano-objects sensitized ones. It permits to conclude the following main idea of this brief analysis: the existing of the innovative tendency to use the bio-objects instead the nano-objects ones to optimize the organics photorefractivity.

In order to predict the practical application of this study one can consider the use of the nano- and the bio-objects doped compounds in the electrically- and optically-addressed spatial light modulator construction, which have been classically applied for the correction of the phase aberrations, for the conversion of the laser beams, for the switching, for the limiting, for the surface-plasmon generation, etc. [61–69]. In any case, the refractive properties of structures are very important.

According of the structures shown in this paper, really, it should be noticed that for the UV spectral range the ZnS, ZnSe photolayers can be considered; for the VIS spectral range the pure polyimide, COANP, LC films can be applied, but for the near-IR spectral range the nano- and bio-objects doped studied systems can be proposed with good advantage. Thus, the spectral and energy range of the practical application of the materials studied can be effectively predicted.

4. Conclusions

To conclude this consideration it can be testify the following main facts:

The innovative tendency to reveal the advantage of the bio-objects sensitization instead the nano-objects ones has been accented and supported. Bio-objects-doped structures shown the same or the better refractive parameters that have been previously observed for the nano-object-doped ones. Bio-objects-doped organic matrixes can be used with good advantage due to the reason that they are renewable and the non-toxic systems. The photorefractive parameters change of the organics via the nano- and the bio-structuration process can be considered as the indicator of following dynamic and photoconductive characteristics modification. The results obtained can extend the perspective using of the nano- and the bio-objects-doped organic composite materials in the general optoelectronics and biomedicine as well as. Specific area of the modulation and holographic technique can be extended via the application of the nano- and bio-objects doped systems due to spectral and energy range change. Due to the increased absorption cross-section in the nano- and bio-structured materials with the initial CTC formation process, they can be effectively used in the limiter devices in order to protect the human eyes and the technical devices from the high laser irradiation. Due to the easy visualization of the refractivity change effect, it can be used in the education process in order to extend the students′ knowledge in the material science area.

Author Contributions: Conceptualization, validation, formal analysis, writing—original draft preparation, writing—review and editing, N.K.; investigation, S.L.; visualization, Y.Z. All authors have read and agreed to the published version of the manuscript.

Funding: The presented here results are coincided with the main research direction of the Lab for Photophysics of media with nanoobjects and they are the part of a work supported partially by Russian Foundation for Basic Research, grants No.10-03-00916 (2010–2012), No.13-03-00044 (2013–2015) as well as by FP7 Marie Curie International researchers exchange proposal "BIOMOLEC" (2011–2015). Partially this research has been supported by the Innovation Promotion Fund, grant no. 72598/C1-112174.

Data Availability Statement: Not applicable.

Acknowledgments: Some variant of the scheme used to this study can be seen in ref [36]. Kamanina, N.V.; Vasilenko, N.A. Influence of operating conditions and of interface properties on dynamic characteristics of liquid-crystal spatial light modulators. Opt. Quantum Electron., 1997, Vol.29(1), pp.1–9. The author would like to acknowledge her colleagues: F. Kajzar and Ch.Andraud (France), D.P. Uskokovic (Serbia); V.I. Studeonov, P.Ya. Vasilyev, S.V. Serov, student A.A.Kukharchik (Russia) for their help in this study at its different steps.

Conflicts of Interest: The authors declare no conflict of interest.

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